Journal of CO_2 Utilization 30 (2019) 193–204

Contents lists available at ScienceDirect

Journal of CO_2 Utilization

journal homepage: www.elsevier.com/locate/jcou

Sustainability of carbon delivery to an algal biorefinery: A techno-economic and life-cycle assessment

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ARTICLE INFO

Keywords:
Hydrogen carbonate
Cellulosic sugar
Utilization
Microalgae
Biofuel

ABSTRACT

Supplementation of carbon is critical for high productivity cultivation of most microalgae. Moreover, using microalgae for atmospheric CO_2 mitigation to combat climate change is promising as waste sources and atmospheric CO_2 can be utilized to produce useful products. The challenge is developing technologies, processes, and strategies that utilize carbon efficiently such that the overall system is sustainable. Through engineering system modeling combined with techno-economic and life-cycle assessments, this study examined the implications of various delivery methods of carbon to a production-scale algal biorefinery. Five primary carbon sources were considered: atmospheric CO_2; CO_2 from direct chemical or power plant waste emissions; CO_2 that has been concentrated from waste sources and compressed; inorganic carbon in the form of hydrogen carbonate; and organic carbon in the form of cellulosic sugars derived from corn stover. Each source was evaluated assuming co-location as well as pipeline transportation up to 100 km. Sustainability results indicate that economics are more prohibitive than energy and emissions. Of the scenarios evaluated, only two met both the economic and environmental criteria of contributing less than 0.50 US-$ GGE^{-1} and 20 g $CO_{2\text{-eq}}$ MJ^{-1} to the overall system, respectively: uncompressed, pure sources of gaseous CO_2 with pipeline transportation of 40 km or less; and compressed, supercritical CO_2 from pure sources for pipeline transportation up to 100 km. A first order scalability assessment of algal biofuels based on these results shows carbon to be a limiting nutrient in an algal biorefinery with a total US production capability of 360 million gallons of fuel per year.

1. Introduction

The prevalence of fossil-derived fuels to power the world's modern energy system, including coal and natural gas for electricity generation and petroleum for transportation, is unsustainable. Quite simply, these resources are finite, and cannot support the global human population indefinitely. A more immediate concern, however, is severe and rapid global climate change due to the atmospheric accumulation of carbon dioxide (CO_2) and other greenhouse gases primarily from the burning of fossil fuels. This sobering reality has prompted researchers to investigate alternative fuels and CO_2 mitigation strategies in an effort to decelerate and avert lasting consequences.

One such area of investigation is microalgae. Like terrestrial plants, most microalgae fix carbon from CO_2 during photosynthesis. However, microalgae have the advantage with regard to CO_2 fixation, as they utilize it at a higher rate and supplementation can be controlled [1,2]. Using microalgae to combat climate change is promising, as waste sources and atmospheric CO_2 can be utilized to produce useful products such as renewable fuels and bio-based chemicals and products [3,4]. The current challenge is the holistic development of strategies, technologies, and processes to improve the sustainability of utilizing CO_2 for microalgae cultivation [5–7]. Currently, CO_2 does not represent a limiting resource, but efficient delivery and utilization of CO_2 is critical for large-scale microalgae systems. Various methods of carbon supplementation have been explored, with direct injection of CO_2 enriched gas predominately used. Carbon can also be delivered through hydrogen carbonate solutions, cellulosic sugars or dissolved carbon in urban wastewaters [1,2,8–13]. However, the sustainability of delivering carbon by these methods remains underexplored.

Although carbon is a requisite nutrient for the growth of microalgae it is often neglected in sustainability assessments [14,15]. Most assessments to-date assume co-location with an industrial point source of waste CO_2 without consideration to siting or scalability. Typical sources of CO_2 considered for algal cultivation are waste streams from power or

Abbreviations: TEA, techno-economic assessment; LCA, life-cycle assessment; MFSP, minimum fuel selling price; GGE, gallon of gasoline equivalent; GWP, global warming potential; NER, net energy ratio; WTW, well-to-wheels

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https://doi.org/10.1016/j.jcou.2019.01.007
Received 6 September 2018; Received in revised form 14 January 2019; Accepted 24 January 2019
Available online 21 February 2019
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chemical manufacturing plants. These sources of CO_2 range in concentration from low, approximately 3–5 wt% and 12–15 wt% from natural gas and coal-fired power plants, respectively, to high, at 99 wt% from ethanol plants or other chemical plants, like ammonia or hydrogen [16]. However, even if co-location is assumed, the additional cost and energy to transport and deliver CO_2 on-site are typically absent; rather, the assumption implies that co-location constitutes a free source of CO_2. Some studies have considered the implications of on-site gaseous delivery with results showing a significant impact; specifically, up to 20–25% of the operational costs of the growth system are attributed to on-site gaseous carbon delivery [16–23]. The failure to integrate CO_2 delivery on-site or further transport to the growth facility represents a severe oversight.

Proposed options for CO_2 transportation and delivery to algal biorefineries have included pipelines or commercially available, bottled CO_2 [18]. The practicality of bottled CO_2 for large-scale production is questionable, so pipeline transportation is considered the only viable option. Further, pipeline options include transportation of flue gas at low or high pressures, or transportation of pure, supercritical CO_2. The concentration of the CO_2 source will influence the pipeline design and the distance over which CO_2 may be cost-effectively transported [16]. There is a tradeoff between the capital cost of a pipeline and its power consumption, the latter of which increases operational costs and negatively impacts life-cycle metrics [18]. For example, it may be more cost-effective to transport pure CO_2 in terms of capital expenses (smaller volume, therefore smaller pipeline), but at the cost of greater power consumption [16]. The distance over which CO_2 may be cost-effectively transported will influence siting options and resource assessments, as fewer point-sources may be suitable due to surrounding land availability. In general, low-pressure transportation over short distances is preferred, but not necessarily practical, as the short distance limits cultivation siting and the size of the cultivation facility [19]. Previous studies have attempted to address the question of the economical transport distance of CO_2 from industrial point sources, as well as comparison to the purchase of commercially available CO_2. The earliest assessments were completed by Benemann et al. [24] and Kadam [25]. Benemann et al. [24] concluded that the maximum economical distance of CO_2 transported as low-pressure flue gas in 2-meter diameter pipes was 5 km. Kadam [25] evaluated the cost difference between transporting flue gas versus purified, or concentrated CO_2, over 100 km, and found that for a 500 MW coal-fired power plant, the cost of delivery for purified CO_2 was 40.50 US-$ metric ton^{-1} and the cost for direct flue gas was 57.20 US-$ metric ton^{-1}. Based on this early work, some recent publications have attempted to integrate considerations of CO_2 transport, delivery, or cost, but are limited to very specific case studies and are often underreported [16–20,22,26–32]. To feasibly scale any proposed algal biorefinery, a thorough understanding of carbon resource transportation and delivery is critical.

Another consideration for CO_2 cost and delivery is utilization efficiency. In addition to productivity and facility size, the CO_2 utilization efficiency, or the efficiency at which microalgae consumes available CO_2 during growth, directly impacts the volume demand of CO_2 delivered to an algal cultivation facility [16]. Many microalgae-based biofuel sustainability assessments assume greater than 75% of the CO_2 is utilized or recovered in the biomass with no validated experimental evidence [20,21,26,29,33,34]. Some authors have even suggested that the efficacy of CO_2 injection into algal bioreactors may be significantly less than proponents have claimed [1,15]. Although CO_2 is not currently a limiting resource, overestimation of carbon utilization efficiency may result in the underprediction of economic feasibility and environmental impacts.

The primary objective of this work was to explore the sustainability implications, both economic and environmental, of delivering carbon to a production-scale algal biorefinery. Techno-economic assessment (TEA) and life-cycle assessment (LCA) were applied to a supporting engineering process model to estimate the effective cost per carbon in the produced biomass, minimum fuel selling price (MFSP), global warming potential (GWP) and net energy ratio (NER) for multiple carbon delivery scenarios. The carbon sources considered included atmospheric CO_2; CO_2 from direct chemical or power plant waste emissions; CO_2 that has been concentrated from waste sources and compressed; inorganic carbon in the form of hydrogen carbonate or bicarbonate salt; and organic carbon in the form of cellulosic sugars derived from corn stover. Each source was evaluated assuming co-location as well as pipeline transportation up to 100 km. Discussion focuses on solutions that were proven to be both economically and environmentally viable defined by meeting a cost metric of 0.50 US-$GGE^{-1} and environmental metric of 20 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively, sensitivity to transportation distance, investigation of the impact of utilization efficiency, and the implications of results on the scalability of algal systems based on resource availability.

2. Methods

TEA and LCA require an engineering process model to serve as the foundation for assessment. For this study, a modular engineering process model was developed from literature to include all sub-processes of microalgae cultivation, harvest and conversion to biofuel, as well as upstream models for carbon transportation and delivery. The modularity of the model facilitates evaluation of the alternative carbon delivery scenarios. The primary products are fuel (diesel and naphtha) with co-products of excess electricity from the combined heat and power system and concentrated nutrients recycled to the growth system from the thermochemical conversion process. The following sections outline the assumptions and equations that underlie the engineering process model, detail the carbon sources evaluated in this study, and present the TEA and LCA methods and metrics.

2.1. Engineering process model

The engineering process model developed for this study included all relevant sub-processes for a Well-to-Wheels (WTW) system boundary, including cultivation, harvest/dewatering, conversion and upgrading, a combined heat and power (CHP) plant, transportation and distribution of the biofuel product (T&D), and combustion. A complete process flow diagram and table of key sub-process model parameters is presented in the supplementary material.

2.1.1. Cultivation

The model assumes cultivation of *Galdieria sulphuraria* in 4 hectare (10-acre) open-raceway ponds circulated by paddlewheels and characterized based on pond models described by Davis et al. [26] and Stephenson et al. [35]. The pond circulation energy was estimated to be 1.42 kW ha^{-1}. For the baseline, pure CO_2 was assumed to be delivered via gas sparging and was estimated to require approximately 0.08 kW ha^{-1}. This energy requirement would increase for sparging more dilute sources of CO_2 like power plant flue gas. Energy for pumping water and culture to and from harvesting, respectively, was estimated to be approximately 0.6 kJ kg^{-1}, and off-site make-up water delivery was estimated to be approximately 1 kJ kg^{-1} water. To enable valid comparisons between inorganic and organic carbon sources, *G. sulphuraria* was selected as it can be grown phototrophically, heterotrophically, or mixotrophically [10,12]. It has a carbon, nitrogen, and phosphorus (C:N:P) ratio of 91:17:1 and an assumed composition of 8.9%, 53.3%, and 18.1% lipids, protein, and carbohydrates, respectively (Lammers, personal communication, manuscript in preparation; [33]). Nitrogen and phosphorus were assumed to be supplied by ammonia and diammonium phosphate (DAP) [26]. Baseline productivity and cultivation area were assumed to be 25 g m^{-2} day^{-1} and 2020 ha (5000 acres) to achieve a constant biomass yield of 506 metric tons day^{-1} [26,37]. The total land area for the facility is 3080 ha (7615 acres) and includes infrastructure requirements corresponding to

a packing factor of approximately 0.66 [26]. Seasonality and other temporal effects of productivity were not considered in this study, but the potential implications are discussed. Combined baseline cultivation energy requirements, including pond circulation, CO_2 sparging, and on-site circulation of water and other material streams was estimated to be approximately 1.2 kJ kg^{-1} AFDW algae.

2.1.2. Harvest/dewatering

A three-step harvest and dewatering process was assumed to concentrate the algae from 0.5 g L^{-1} to 20 wt% ash-free dry weight (AFDW), which is suitable for downstream thermochemical conversion [26,38]. This three-step process comprised bioflocculation, dissolved air flotation (DAF), and centrifugation [26,32,34,39]. Combined harvest energy requirements were estimated to be approximately 0.73 kJ kg^{-1} AFDW algae. Algal losses and separated water are assumed to be recycled to the ponds in the cultivation stage at a net algal separation efficiency of 81%.

2.1.3. Conversion and upgrading

The downstream conversion process of microalgae to fuel products was assumed to be hydrothermal liquefaction (HTL) with upgrading through hydrotreating modeled based on Jones et al. [38]. Products of HTL include bio-oil, solids, an aqueous phase, and off-gas (primarily methane). Based on experimental data for the conversion of *G. sulphuraria* via HTL, a bio-oil yield of 35% AFDW algae was assumed (Lammers, personal communication; [36]). The bio-oil is further upgraded by hydrotreating and hydrocracking to diesel and naphtha fuel. The composition of the bio-oil from HTL is assumed to be similar to that of Jones et al. [38]. For this study, it was assumed that the hydrogen for the hydrotreating process was purchased commercially and supplied at a rate of 435 kg h^{-1}. The aqueous phase is processed through catalytic hydrothermal gasification (CHG), during which additional off-gas is produced. The nutrient-laden water is then assumed to be recycled to the ponds in the cultivation stage [40,41]. The recycle of these nutrients decreases the raw inputs to the system with this captured as a part of the foundational engineering process model. The produced gas from HTL, CHG, and hydrotreating is assumed to be sent to a co-located CHP plant to generate on-site electricity and heat [28,42]. Combined electricity and heat requirements for downstream conversion were estimated to be approximately 3 kJ kg^{-1} AFDW algae, and recoverable energy from CHP was estimated to be approximately 3.3 kJ kg^{-1} AFDW algae.

To complete the system, transportation and distribution of the fuel product was included and considers proportions of fuel transported to a terminal by barge, rail, and heavy-duty truck, then further transported to individual stations by truck [28,43]. Energy requirements for transportation and distribution were estimated to be approximately 0.06 kJ kg^{-1} AFDW algae. Emissions from combustion of the fuel product as well as emissions credits are discussed separately in Section 2.5.

2.2. Carbon Sources

This study considered five primary inorganic and organic carbon sources and included both point sources and processing, as well as transportation to an algal facility, Fig. 1. The five primary sources were atmospheric CO_2; CO_2 from direct chemical or power plant waste emissions; CO_2 that has been concentrated from waste sources and compressed; inorganic carbon in the form of hydrogen carbonate salt; and organic carbon in the form of cellulosic sugars derived from corn stover. Each source of carbon is described in greater detail below, followed by its respective transportation method. It was assumed for all cases that carbon is supplied to the ponds 12 hours day^{-1}, representing algal growth only during the daylight hours.

2.2.1. Atmospheric CO_2

Algae may be grown on atmospheric CO_2, but because of its low concentration in air (400 ppm) and mass transfer potential, algal productivity may be up to 90% lower than that of a higher concentration of CO_2 supplemented by gas sparging [1,2]. For the case of growth on atmospheric CO_2, the productivity was assumed to be 2.5 g m^{-2} day^{-1}, an order-of-magnitude lower than the baseline. To maintain a constant biomass yield of 506 metric tons day^{-1}, the facility area was increased to 30,800 ha (76,120 acres). One advantage of growth on atmospheric CO_2 is that no additional provisions are required to supply carbon to the algae. Thus, transportation and utilization efficiency are excluded in the analysis.

2.2.2. Direct waste emissions CO_2

Point sources considered for waste CO_2 emissions included natural gas and coal power plants, as well as ethanol, ammonia, and hydrogen plants [16]. Emissions from natural gas power plants contain 3–5 wt% CO_2, emissions from coal power plants contain 12–15 wt% CO_2, and ethanol, ammonia, and hydrogen plants emit waste streams of CO_2 at greater than 99 wt% concentration. Algal productivity on these waste emissions of CO_2 was assumed to be equal to the baseline of 25 g m^{-2} day^{-1}. The disadvantage of using plant waste emissions directly, specifically dilute sources like natural gas and coal power plant flue gases, is that transporting and delivering large volumes of gas to the algal ponds is challenging and cost intensive [26]. For these cases, it was assumed that the gas is transported by pipeline both off-site and on-site at low-pressure and delivered to the algae ponds via gas sparging. Depending on the sparging method, the utilization efficiency may range from 20% to 90% [1,26,29,44]. The utilization efficiency assumed for the primary cases was 90%, followed by a sensitivity of utilization efficiencies from 10% to 100%.

2.2.3. Captured CO_2

CO_2 from these same point sources (and including atmospheric CO_2) may alternatively be concentrated and compressed to a supercritical state prior to transportation. A common method to capture CO_2 from the atmosphere and power plant emissions is using a monoethanolamine (MEA) solution that is then heated and regenerated to release a nearly pure stream of CO_2 [45]. Algal productivity on these pure streams of captured CO_2 was assumed to be equal to the baseline of 25 g m^{-2} day^{-1}. Although it is more energy intensive to compress CO_2, in a supercritical state it can be readily transported and in smaller pipelines, as it has liquid-like properties. For these cases, it was assumed that the CO_2 is transported by pipeline at supercritical pressures and delivered to the algae ponds via gas sparging. The utilization efficiency assumed for the primary cases was 90%, followed by a sensitivity of 10–100%.

2.2.4. Hydrogen carbonate

Algae may also be grown on CO_2 that has been captured in the form of soluble hydrogen carbonate salts [2,9,13]. However, algal growth on hydrogen carbonate requires a high pH environment [9]. It is unknown if *G. sulphuraria* may be grown on hydrogen carbonate but was assumed for the purposes of this study. Sodium hydrogen carbonate was selected for this study, as it is one of the more common and inexpensive hydrogen carbonate salts. Sodium hydrogen carbonate is commercially produced from sodium carbonate (soda ash), which is mined from trona ore, and when dissolved in water may be used as a sorbent to capture CO_2 similar to the MEA process previously mentioned, forming a hydrogen carbonate solution [46,47]. Algal productivity on hydrogen carbonate was assumed to be equal to the baseline of 25 g m^{-2} day^{-1}. It was also assumed that the hydrogen carbonate was transported by pipeline and delivered to the algae ponds as an aqueous solution. Since the carbon is dissolved, the utilization efficiency was assumed to be 100%.

2.2.5. Cellulosic sugar

Some algae may be grown heterotrophically or mixotrophically on

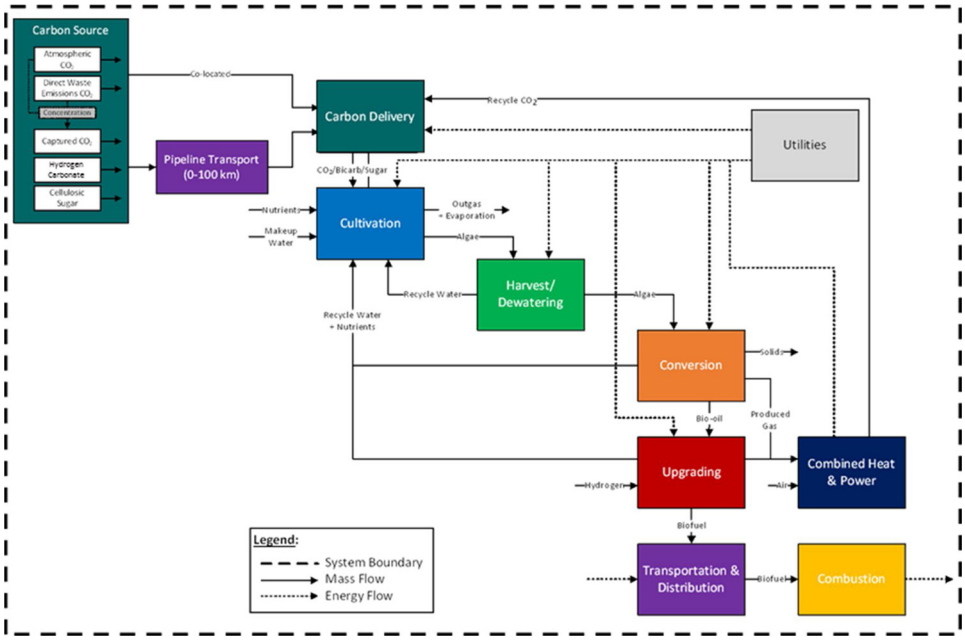

Fig. 1. Carbon Delivery and Engineering Process Model. Flow diagram illustrating the carbon delivery cases evaluated in this study. Organic and inorganic sources of carbon were considered, including optional processing to concentrate a dilute CO_2 source, transportation to an algal facility via pipeline and algal facility distribution. This carbon delivery model was coupled with a traditional engineering process model for an algal biorefinery.

organic carbon such as cellulosic sugars. It has been demonstrated that *G. sulphuraria* may be grown both heterotrophically and mixotrophically, so a case was considered for enhanced mixotrophic growth on cellulosic sugars derived from corn stover [12,48]. In mixotrophic growth, a proportion of the microalgae growth is phototrophic on CO_2 and the remaining is heterotrophic growth on sugars. Previous work on heterotrophic growth of *G. sulphuraria* on pure sugars yielded a volumetric productivity of 1.08–1.15 g L^{-1} day^{-1}, with a substrate yield (mass ratio of algae biomass to input sugar) of 0.48–0.50 [12]. Based on experimental trials at Arizona State University, mixotrophic algal productivity was assumed for this case to be 1.37 g L^{-1} day^{-1} (equivalent to approximately 91 g m^{-2} day^{-1}) with a substrate yield of 0.97 (Lammers, personal communication). The increase over previously demonstrated heterotrophic productivity is reflective of the additional phototrophic growth. It should be noted that this productivity was demonstrated indoors and in closed, glass tubular photobioreactors, therefore the assumption that this productivity could be achieved in outdoor open-raceway ponds is for the purposes of this study only. At this enhanced productivity, the facility area was decreased to 850 ha (2090 acres) to maintain a constant biomass yield of 506 metric tons day^{-1}. As with the hydrogen carbonate case, it was assumed that the cellulosic sugar was transported by pipeline and delivered to the algae ponds as an aqueous solution. The utilization efficiency was also assumed to be 100%.

2.3. Carbon capture and transportation models

Models were developed to characterize the properties, processing, and transportation of the carbon sources considered. Parameters characterizing this modeling may be found in the supplementary material. Critical to the modeling of CO_2 capture and transportation are accurate fluid properties as they change as the gas deviates from ideal behavior. Power plant waste emissions were modeled as gas mixtures of air and 4 wt% and 13.5 wt% CO_2 for natural gas and coal, respectively. Density, viscosity, compressibility factor, and specific heats for these gas mixtures and pure CO_2 were tabulated using NIST REFPROP for appropriate pressures and temperatures [49].

2.3.1. Carbon capture

The carbon capture method considered in this study was monoethanolamine (MEA) scrubbing. In this process, CO_2 gas mixtures are passed through an absorber with MEA solvent to capture the CO_2. The solvent is then heated and regenerated to release the captured CO_2 as a pure stream. This is an energy-intensive process, and the first and second laws of thermodynamics define the minimum work required for CO_2 separation. Also, the work required for separation increases as the concentration of CO_2 in the gas mixture decreases [45]. To estimate the energy for separation of CO_2 from the atmosphere and power plant waste emissions, a formulation of minimum thermodynamic work as a function of gas mixture molar fractions combined with an empirical estimate of second-law efficiency was applied [45]. For atmospheric CO_2 capture (400 ppm), capture from a natural gas power plant (4 wt %), and capture from a coal power plant (13.5 wt%), energy requirements were estimated to be 34.4, 1.4, 0.5 MJ $(kg\ CO_2)^{-1}$, respectively. Combined capital costs for carbon (CO_2) capture and compression equipment were proportionally scaled from costs presented in National Energy Technology Laboratory reports for similar systems [50]. The equipment cost, including capital and installation, was estimated to be 38,361 US-$ metric ton^{-1} CO_2 captured per day with scaling factor information presented in the SI.

2.3.2. Compression

Once separated, CO_2 may be compressed and cooled for pipeline transportation as a supercritical fluid. Pipeline transport conditions are typically 10–15 MPa and 35 °C [45,51]. Gas compression to these pressures results in increased temperature, which may exceed the material limits of a compressor. Therefore, compression occurs in multiple stages with cooling. In the model developed for this study the maximum compressor temperature was 150 °C and the maximum compression ratio per stage was 6 [52]. The temperature increase for each compression stage was determined using isentropic relations for temperature and pressure, and a compression ratio and number of stages was determined as to not violate the previous criteria. Between each compression stage, the gas was cooled to 25 °C. Energy requirements for compression and cooling were based on equations for isentropic compression and basic heat work, respectively [45,51,52]. For compression to 14 MPa with inter-stage cooling, the energy requirement was estimated to be 1 MJ $(kg\ CO_2)^{-1}$. As indicated above, combined capital costs for carbon (CO_2) capture and compression equipment were estimated to be 38,361 US-$ metric ton^{-1} CO_2 captured per day [50].

2.3.3. Transportation

Pipeline transportation was modeled for all carbon sources and cases, except for growth on atmospheric CO_2. A maximum transport distance of 100 km was evaluated based on previous evaluations that show this to be a realistic upper bound. The majority of assessments surveyed assume a pipeline distance of less than 5 km [22,24,53]. Select studies investigated distances larger than this with an upper bound of approximately 100 km [23,25]. The primary design parameter of a pipeline is diameter, which must be sized for a given pressure and flow rate, which is further a function of facility carbon demand. For this study, a steel pipeline was modeled based on characteristic equations for internal compressible flow, accounting for fluid compressibility and friction losses [45,51,52]. Losses due to elevation change or bends were neglected. This formulation is iterative and requires a target pressure drop and an initial estimate of pipeline diameter based on a desired flow velocity for a given mass flow rate, which varies for each carbon source (see supplementary material). The model solves for a pipeline diameter to meet the specified criteria which is then adjusted to a standard nominal pipe size, as described by McCoy and Rubin [51].

Depending on pressure losses and properties of the fluid to be transported, intermediate booster compressor stations may be required to re-pressurize the fluid for transportation over long distances. Similarly, multiple pipelines running in parallel may be required to accommodate a large fluid volume. The model accounts for both scenarios by limiting the pipeline diameter to 2 m and adding a booster station or parallel pipeline if the diameter exceeds that limit to accommodate a large pressure drop or fluid volume, respectively. Energy requirements for booster stations were based on equations for isentropic compression or pumping for gaseous and liquid (or supercritical) transport, respectively. Booster stations are required for direct waste emissions from natural gas and coal power plants, which added up to an estimated 247.2 and 66.2 kW km^{-1}, respectively, to overcome pressure losses for a 100 km pipeline. Aqueous transportation of hydrogen carbonate (100 g L^{-1}) and cellulosic sugar (127 g L^{-1}) require an estimated pumping power of 21.9 and 12.8 kW km^{-1}, respectively.

Capital costs—including materials, labor, right-of-way, and miscellaneous other costs—in addition to operation and maintenance costs, were based on a steel pipeline cost model for the Midwest region [51]. Pipeline equipment capital and installation costs for transporting direct waste emissions from a natural gas power plant, coal power plant, and pure sources of CO_2 were estimated to be 33.2 M (million) US-$, 5.3 M US-$, and 1.6 M US-$ km^{-1}, respectively. These dilute and low-pressure sources of CO_2 require significant pipeline infrastructure and multiple pipelines to accommodate the large gas volumes. Alternatively, liquid transportation of captured and compressed CO_2 was estimated to cost 197 k US-$ km^{-1} in equipment capital and installation. Aqueous transportation of hydrogen carbonate and cellulosic sugar were estimated to cost 405 k US-$ and 184.6 k US-$ km^{-1}, respectively.

2.3.4. On-site distribution

Three methods for the on-site distribution of carbon were considered for low-pressure gas, pressurized CO_2, and aqueous carbon. Davis et al. [26] describes two designs for the distribution of CO_2. The first design assumes distribution of CO_2-enriched gas from direct waste sources like natural gas or coal power plants using fans and blowers. The second design assumes liquid storage of pure CO_2 which is slowly heated and vaporized to distribute throughout the facility at moderate pressure. The energy requirements were estimated to be 37.1 and 142.6 kJ (kg gas)$^{-1}$, respectively. Equipment capital and installation costs were estimated to be 9974 US-$ and 4783 US-$ (kg gas h^{-1})$^{-1}$, respectively. Both hydrogen carbonate and cellulosic sugar were assumed to be delivered as aqueous solutions and were distributed on-site with make-up water delivery, requiring no additional energy, capital costs, or other provisions.

2.4. Techno-economic assessment

TEA for this study was based on the costs reported by Davis et al. [26] for upstream systems (cultivation and harvest/dewatering), with downstream costs based on the work of Jones et al. [38] and Schwab et al. [37]. All costs were converted to 2014 U.S. dollars such that direct comparison to existing literature could be made. The capital and operating costs drawn from these studies were adjusted to account for differences in the overall process modeled. Capital costs of equipment for on-site CO_2 distribution and downstream processing were scaled using appropriate exponents and relevant material mass flow rates with details presented in the SI [54]. Scaling was required due to the lower bio-oil yield of *G. sulphuraria* for downstream processing. Equipment costs for on-site hydrogen production were also removed, and hydrogen was added as a material input. Other operating costs were scaled proportionally with mass flow rate, and included nutrients (ammonia and DAP), electricity, and natural gas. CO_2 feedstock costs at 45 US-$ metric ton^{-1} were removed entirely for the baseline model to be replaced with the different scenarios considered in this study. Costs included direct and indirect capital investment for the algal facility and any carbon delivery systems, land, variable operating costs of raw materials and energy inputs, fixed operating costs of labor, maintenance, and insurance, and co-product credits for excess electricity sold back to the grid. Costs for select cases are outlined in the supplementary material.

Capital costs, operational costs, and yield of the system were input into a 30-year discounted cash flow rate of return (DCFROR) analysis. Nth-plant assumptions were considered for the DCFROR [26,38,55], which most notably assumes a discount rate (equal to an internal rate of return for this work) of 10%, a 35% tax rate, and 330 days of operation per year for 30 years. The assumed lifetime of the system is based on the expected life of the system [26,27,56,57]. The DCFROR analysis outputs a minimum fuel selling price (MFSP) for a functional unit of 1 gallon of gasoline equivalent (GGE), which was calculated for the diesel and naphtha fuel products using ratios of heating values. For individual cases, the difference in MFSP relative to the baseline constituted the effective cost in net-present-value (NPV) for the defined transportation and delivery of the given carbon source. Using the MFSP difference, biomass and fuel production mass flow rates and mass flow rate of carbon input, the cost of carbon was calculated on a per mass basis of the carbon feedstock, as well as on a normalized, per carbon basis.

2.5. Life-cycle assessment

The mass and energy balance calculated from the engineering process model supported the LCA work. The output of the LCA was the environmental impact of the system quantified by a global warming potential (GWP) and net energy ratio (NER) of the process. The functional unit for both GWP and NER was 1 MJ of fuel product.

Life-cycle inventory (LCI) data from various public databases and literature sources was gathered for all raw material and energy inputs. This data, combined with the 100-year global warming equivalence factors of 1, 34 and 298 for carbon dioxide (CO_2), methane (CH_4) and dinitrogen oxide (N_2O), respectively, were used to calculate the GWP in g $CO_{2\text{-eq}}$ MJ^{-1} [58]. Emissions credits were applied for the carbon captured in the produced fuel and HTL solids, as well as for the electricity and natural gas displaced by on-site CHP. Emissions from combustion of the fuel were included for the WTW system boundary.

In additional to GWP, NER was calculated for the process. NER is defined as the ratio of direct energy input to direct energy output of the process. Direct energy inputs represent process consumption of electricity and natural gas for heat, minus credits for on-site generation via CHP. The direct energy output is the energy content of the produced fuel based on its heating value. An NER of less than one is typically considered favorable.

3. Results and discussion

A modular engineering system model was developed and validated from literature sources for an algal biorefinery. Coupled to this model was the development of various carbon feedstocks and a pipeline transportation and delivery model. Modularity supported the evaluation of various alternative carbon delivery pathways including atmospheric CO_2, CO_2 from direct chemical or power plant waste emissions, CO_2 that has been concentrated from waste sources and compressed, inorganic carbon in the form of hydrogen carbonate salt, and organic carbon in the form of cellulosic sugars derived from corn stover. The engineering system model was coupled with economic and environmental impact modeling. TEA and LCA results were used to evaluate the different carbon delivery methods compared to a baseline scenario with regard to carbon feedstock cost, fuel selling price, process emissions, and process energy. Sensitivity to transportation distance and carbon utilization efficiency, as well as other resource considerations are discussed.

3.1. Baseline scenario results

All cases were evaluated relative to a common baseline, which assumes that the carbon feedstock is pure CO_2 that has been captured and stored as a liquid in a pressurized storage system, with no associated economic or energetic cost aside from the energy allocated to on-site delivery of pure CO_2 to the ponds at 90% utilization efficiency [26]. The baseline MFSP, GWP, and NER were calculated to be 6.47 US-$ GGE^{-1}, 28.1 g $CO_{2\text{-eq}}$ MJ^{-1}, and 0.13 MJ MJ^{-1}, respectively. Figures and breakdowns by cost category or sub-process may be found in the supplementary material. By reporting results relative to a common baseline case, scenarios may be compared directly to understand the differences in economic and environmental impacts.

3.2. Cost of carbon

Prices of carbon feedstocks on the market are typically given on a mass basis of the feedstock itself. Prices may fluctuate marginally with production volume according to basic economic principles, but in general and for the purposes of this study, feedstock prices stay relatively fixed. However, a kilogram of CO_2, hydrogen carbonate, and cellulosic sugar are not equivalent for similar applications, as the mass percentage of carbon contained in each of those feedstocks differs. As such, the advertised feedstock prices on a mass basis cannot be directly compared. Furthermore, in the case of microalgae cultivation, the utilization efficiency of a carbon resource, as well as any recycling of carbon within the system, will influence the effective feedstock cost as part of the production process, even if the feedstock price is unchanged. Fig. 2 presents the effective costs for each carbon source considered in this study on a per mass basis of the carbon feedstock (yellow), as well as on a normalized, per carbon basis (purple). As utilization efficiency decreases, the effective cost of carbon when applied to the production process will increase.

Costs in Fig. 2 are reported as effective costs, which account for off-site transportation costs (if not co-located), on-site provisions for distribution to the ponds, any capital and operational cost tradeoffs for the facility based on productivity, and raw material purchase costs for the carbon feedstock, all levelized by calculating the NPV of the 30-year cashflow. On a carbon basis, the highest effective costs (greater than 400 US-$ metric ton^{-1} carbon) were associated with growth on atmospheric CO_2; utilization of direct emissions from a natural gas or coal power plant with 100 km of pipeline transportation; captured and pressurized CO_2 from the atmosphere; and hydrogen carbonate. Mid-range effective costs (between 150 US-$ and 400 US-$ metric ton^{-1} carbon) were associated with direct emissions from a co-located natural gas power plant; direct emissions from a pure CO_2 source with 100 km of pipeline transportation; captured and pressurized CO_2 from a natural gas or coal power plant; and cellulosic sugar. The lowest effective costs (less than 150 US-$ metric ton^{-1} carbon) were associated with utilizing direct emissions from a co-located coal power plant or pure CO_2 source; and captured and pressurized CO_2 from a pure source. Because direct waste streams of CO_2 are dilute (except pure CO_2 sources) and transported at low-pressure, the volume to be transported is much greater, so transportation costs for all direct waste streams of CO_2 were significantly higher than all other cases. Transporting compressed CO_2 as a supercritical fluid by pipeline over 100 km added 11.50 US-$ metric ton^{-1} of CO_2, whereas the lowest cost to transport a direct emissions source at low-pressure was 86.50 US-$ metric ton^{-1} corresponding to a pure source, with costs increasing for coal and natural gas flue gas. These results demonstrate that low-pressure transport represents a significant challenge for gaseous carbon delivery.

All cases were evaluated for an algal productivity of 25 g m^{-2} day^{-1}, with two exceptions: growth on atmospheric CO_2 was assumed to be an order-of-magnitude less at only 2.5 g m^{-2} day^{-1} and mixotrophic growth on cellulosic sugars was assumed to be 91 g m^{-2} day^{-1}. In the case of atmospheric CO_2 there is no raw material purchase cost, as the CO_2 diffuses from ambient air, but due to the order-of-magnitude increase in facility area and capital costs to compensate for the low algal productivity (while maintaining a fixed biomass yield of 506 metric tons day^{-1}), there is an effective cost of the carbon relative to the baseline of 1806 US-$ metric ton^{-1} of CO_2. Conversely, mixotrophic growth on cellulosic sugars increases productivity by more than 3-fold, which results in a significant decrease in

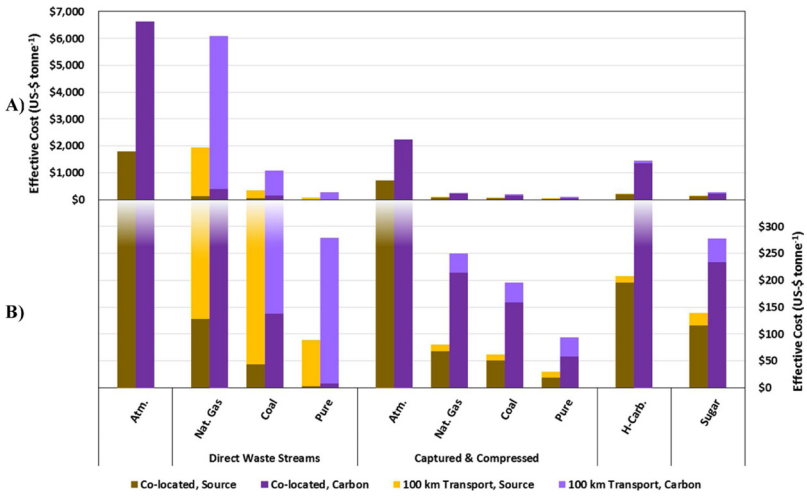

Fig. 2. Carbon cost results. Effective cost in 2014 US-$ of the carbon sources evaluated in this study. Results are shown in two panels: (A) full range of results, (B) results less than 350 US-$ metric ton^{-1} (*Atm*, atmosphere; *Nat. Gas*, natural gas; *H-Carb*, hydrogen carbonate). Sources include gaseous CO_2, hydrogen carbonate, and cellulosic sugars derived from corn stover. CO_2 sources may be in the form of a direct waste stream like power plant flue gas or captured CO_2 that is concentrated and compressed. Results are presented on a mass (metric ton) basis of each respective carbon source (yellow), as well as on a mass basis of carbon in the resulting biomass (purple). Transportation distances are presented for co-located sources and 100 km of added transport via pipeline. All cases assume 90% utilization of the carbon by the microalgae, except for hydrogen carbonate and sugar which assume 100%.

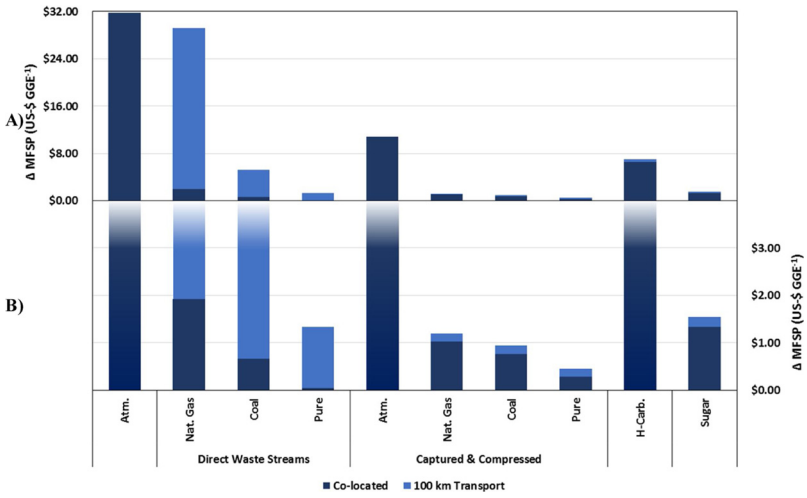

Fig. 3. Fuel price results. Minimum biofuel selling price (MFSP) in 2014 US-$ per gallon of gasoline equivalent (GGE) for the carbon sources evaluated in this study. Reported as a difference relative to the baseline MFSP of 6.47 US-$ GGE^{-1}. Results are shown in two panels: (A) full range of results, (B) results less than 4 US-$ GGE^{-1} (*Atm.*, atmosphere; *Nat. Gas*, natural gas; *H-Carb.*, hydrogen carbonate). Transportation distances are presented for co-located sources and 100 km of added transport via pipeline. All cases assume 90% utilization of the carbon by the microalgae, except for hydrogen carbonate and sugar which assume 100%.

facility area and capital costs at the expense of an increase in variable operating costs from sugar. The raw material purchase price of cellulosic sugars was assumed to be 396 US-$ metric ton^{-1} of sugar, but the effective cost was only 139 US-$ metric ton^{-1} of sugar with 100 km of pipeline transportation. The dramatic increase in productivity offsets the high cost of carbon. The effective cost of hydrogen carbonate changed very little from a raw material purchase price of 200 US-$ metric ton^{-1} of hydrogen carbonate to 208 US-$ metric ton^{-1} with 100 km of pipeline transportation because of minimal energy inputs and no assumed increase or decrease of algal productivity from 25 g m^{-2} day^{-1}. These specific cases illustrate that productivity and associated capital and operational cost tradeoffs may have an influence on the effective cost of carbon source that are not reflected in the raw material purchase cost.

3.3. Carbon sourcing impacts on selling price of fuel

A minimum biofuel selling price (MFSP) was calculated for each carbon feedstock based on the 30-year cashflow. MFSP is presented in Fig. 3 as a difference relative to the baseline result of 6.47 US-$ GGE^{-1}. Fuel selling price is directly proportional to the cost of carbon, with similar trends observed comparatively. However, the allowable cost for the provision of carbon is dependent on the overall process design and fuel price target. If a future target MFSP is 5 US-$ GGE^{-1}, which currently exceeds conventional fuel costs, a 10% allocation for the provision of carbon would be 0.50 US-$ GGE^{-1}. Alternatively, a more conservative estimate of algal based fuels is $3 GGE^{-1} which to achieve is expected to require financial credits for utilization of carbon. In this scenario it is assumed the costs for carbon delivery can be slightly higher, approximately 15% resulting in approximately 0.50 US-$ GGE^{-1} [22,59]. Although it is arbitrary, if the 0.50 US-$ GGE^{-1} threshold is considered in this study, the only carbon feedstock cases that meet that criterion are utilization of direct emissions from a co-located pure CO_2 source and pressurized CO_2 from a pure source with pipeline transportation of 100 km or less.

CO_2 from the atmosphere, natural gas and coal power plants, and carbon delivered in the form of hydrogen carbonate and cellulosic sugar do not meet the 0.50 US-$ GGE^{-1} criterion. CO_2 from the atmosphere is not viable due to the significant decrease in expected algal productivity, or alternatively, the high energy cost to capture and concentrate CO_2 from the atmosphere. For the case of growth on atmospheric CO_2, if the productivity were to remain at 25 g m^{-2} day^{-1}, there would be little expected change in the fuel cost relative to the baseline. This represents an optimal solution for carbon delivery to algal systems but presents a significant technical challenge. CO_2 from a natural gas power plant is not viable due to either the prohibitive capital cost of gaseous transport (both off-site and on-site) or high energy cost to capture and concentrate the CO_2. At a CO_2 concentration of only 3–5 wt%, much of the energy consumption and infrastructure are used to transport or process a substantial proportion of unusable, inert gas. Lastly, CO_2 from a coal power plant is second to pure sources, but it increases the cost of fuel by a minimum of 0.66 US-$ GGE^{-1} for direct waste emissions from a co-located plant. Like natural gas power plants, at a CO_2 concentration of 12–15 wt%, direct waste emissions from a coal power plant require additional energy and infrastructure to transport and process unusable gas.

Although cost penalties for the processing or transportation of large gas volumes are alleviated, neither hydrogen carbonate nor cellulosic sugar proved to be viable based on the 0.50 US-$ GGE^{-1} criterion. The commercial raw material purchase price of hydrogen carbonate at 200 US-$ metric ton^{-1} is cost prohibitive, and unlike cellulosic sugar, growth on hydrogen carbonate is assumed to not increase productivity. The significant productivity gain did lower the effective cost of cellulosic sugar, but the decrease was not significant enough for cellulosic sugar to be viable. However, the offset of capital to operating expenditures and decrease in required land area may still make cellulosic sugar an attractive option if proven at a commercial scale. For both hydrogen carbonate and cellulosic sugar, a significant decrease in raw material prices could improve their viability, but the feasibility of lowering prices was not investigated. Based on the carbon sources considered and their impact on MFSP, results suggest that pure sources of gaseous CO_2 represent the most promising option in terms of economics.

3.4. Greenhouse gas emissions and direct energy usage

The global warming potential (GWP) was calculated for each carbon feedstock based on the metric of CO_2-equivalent ($CO_{2\text{-eq}}$), which incorporates CO_2, CH_4, and N_2O based on their respective 100-year global warming potentials. GWP results are presented in Fig. 4 as a difference relative to the baseline of 28.1 g $CO_{2\text{-eq}}$ MJ^{-1} for algal biorefinery modeled. The allowable $CO_{2\text{-eq}}$ emissions for the provision of carbon is dependent on the overall process design and remaining emissions allocation to meet the Renewable Fuel Standard (RFS) [60]. The RFS is a federal program managed by the U.S. Environmental Protection Agency (EPA) which mandates that advanced fuels, which include microalgae-based fuels, must meet a 50% greenhouse gas (GHG) reduction relative to a 2005 petroleum baseline. On a well-to-wheels (WTW) basis, this 50% reduction is approximately equal to 45 g $CO_{2\text{-eq}}$ MJ^{-1}. Therefore, for this study to meet the RFS, an additional 17 g $CO_{2\text{-eq}}$ MJ^{-1} may be allocated for carbon delivery. Depending on the system this value could go up or down. In this study we evaluate the technologies based on a

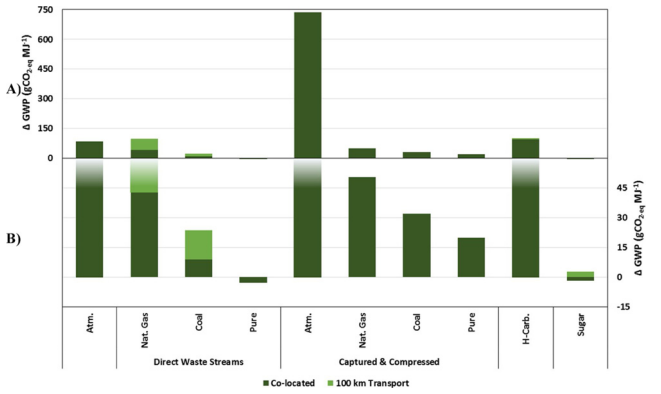

Fig. 4. Process emissions results. Global warming potential (GWP) in grams of CO_2-equivalent per MJ of fuel for the carbon sources evaluated in this study. Reported as a difference relative to the baseline GWP of 28.1 g $CO_{2\text{-eq}}$ MJ^{-1}. Results are shown in two panels: (A) full range of results, (B) results less than 60 g $CO_{2\text{-eq}}$ MJ^{-1} (*Atm.*, atmosphere; *Nat. Gas*, natural gas; *H-Carb.*, hydrogen carbonate). Transportation distances are presented for co-located sources and 100 km of added transport via pipeline. All cases assume 90% utilization of the carbon by the microalgae, except for hydrogen carbonate and sugar which assume 100%.

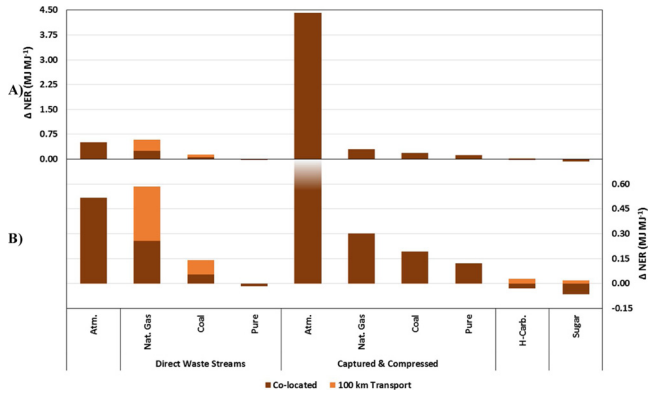

Fig. 5. Process energy results. Net energy ratio (NER) in MJ direct process energy per MJ of fuel for the carbon sources evaluated in this study. Reported as a difference relative to the baseline NER of 0.13 MJ MJ^{-1}. Results are shown in two panels: (A) full range of results, (B) results less than 0.75 MJ MJ^{-1} (*Atm.*, atmosphere; *Nat. Gas*, natural gas; *H-Carb.*, hydrogen carbonate). Transportation distances are presented for co-located sources and 100 km of added transport via pipeline. All cases assume 90% utilization of the carbon by the microalgae, except for hydrogen carbonate and sugar which assume 100%.

threshold of 20 g $CO_{2\text{-eq}}$ MJ^{-1}. Direct waste emissions from a co-located coal power plant; a pure source with 100 km of pipeline transportation; and cellulosic sugar with 100 km of pipeline transportation are the only cases that meet this criterion. However, additional emissions may be allocated to carbon delivery if the baseline process were more energy efficient or included other carbon credits. The allocation for carbon delivery would need to be increased to 20, 24, and 32 g $CO_{2\text{-eq}}$ MJ^{-1} for pressurized CO_2 from a pure source with 100 km of pipeline transportation; direct waste emissions from a coal power plant with 100 km of pipeline transportation; and pressurized CO_2 from a coal power plant with 100 km of pipeline transportation, respectively, to meet the RFS emissions criteria.

It should be noted that based on the assumptions for transportation of a direct, pure CO_2 source at low-pressure, there are no additional emissions as it was found that the outlet pressure at the source is sufficient for transport over the 100 km distance considered. Similarly, no additional emissions are associated with transporting compressed CO_2 as the pressure to bring CO_2 to a supercritical state was found to be sufficient for transportation over the distance considered. Co-location with a direct source of pure CO_2 or a cellulosic sugar production facility were found to lower emissions relative to the baseline, as the provisions for on-site delivery of the carbon are less energy intensive. It should also be noted that with the exception of transporting direct emissions from a natural gas power plant, the GWP associated with capturing and compressing CO_2 exceeds the GWP associated with transporting and delivering direct waste streams of CO_2 at low-pressure. This is because capturing and compressing CO_2 is much more energy intensive and is required even if the point source is co-located in order for the CO_2 to be temporarily stored on-site as a liquid.

CO_2 from the atmosphere or a natural gas power plant and carbon delivered in the form of hydrogen carbonate are not likely to meet the RFS of 45 g $CO_{2\text{-eq}}$ MJ^{-1}. CO_2 from the atmosphere is not viable due to the high energy to capture and concentrate CO_2 from the atmosphere or alternatively, the significant decrease in algal productivity assumed. Similarly, CO_2 from a natural gas power plant is not viable even if co-located due to either the high energy necessary to transport the large gas volume (both off-site and on-site) or high energy to capture and concentrate the CO_2. Lastly, hydrogen carbonate is prohibitive as much of the carbon it contains was extracted from the ground (as soda ash, or sodium carbonate) and would not have otherwise been emitted into the atmosphere (unlike power plant exhaust emissions). This results in a net positive carbon emission as it represents sequestered carbon being released.

Direct energy usage for the overall process was also calculated to determine the net energy ratio (NER), which is presented in Fig. 5 as a difference relative to the baseline of 0.13 MJ MJ^{-1} for the algal biorefinery modeled. The baseline NER is low due to the integration of CHP with the process using biogas (primarily methane) from the downstream conversion process, as well as efficient technologies for upstream cultivation and harvest. For reference, the NER of conventional petroleum diesel is approximately 0.18 [61]. It was previously indicated that an NER of less than one is considered favorable, which allows for an additional 0.87 MJ MJ^{-1} for carbon delivery relative to the baseline. The only scenario in this study that does not meet this criterion is CO_2 captured from the atmosphere, as it is energetically intensive. In general, NER is proportional to GWP, with the exception of hydrogen carbonate and cellulosic sugar, whose emissions are not directly tied to energy consumption but to their feedstocks.

The scenarios with the lowest NER contribution included direct waste emissions from a co-located coal power plant; direct waste emissions from a pure source with 100 km of pipeline transportation; hydrogen carbonate with 100 km of pipeline transportation; and cellulosic sugar with 100 km of pipeline transportation. When co-located, direct waste emissions from a pure source, hydrogen carbonate, and cellulosic sugar were found to reduce the NER, as they are energetically favorable to the baseline. This is because energetic provisions to transport and deliver CO_2 on-site are reduced through low-pressure delivery, or eliminated entirely in the case of hydrogen carbonate and cellulosic sugar, as they are delivered as aqueous solutions with the make-up water. Although NER can be a valuable measure of the viability of energy products like fuels, it was found to be less restrictive than MFSP or GWP in evaluating the sustainability of carbon delivery.

3.5. Sensitivity to transportation distance

Microalgae cultivation is dependent on specific land and siting criteria for scalability and resource considerations [22], so it is critical to evaluate scenarios for which carbon must be transported over potentially long distances to be supplied to an algal facility. In this study, pipeline transportation up to 100 km was considered for delivering direct waste CO_2 emissions from power plants or pure sources, captured and compressed pure CO_2 in a supercritical state, and solutions of hydrogen carbonate and cellulosic sugars. Results for MFSP and GWP are presented in Fig. 6 as differences relative to the baseline of 6.47 US-$ GGE^{-1} and 28.1 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively, for transport distances

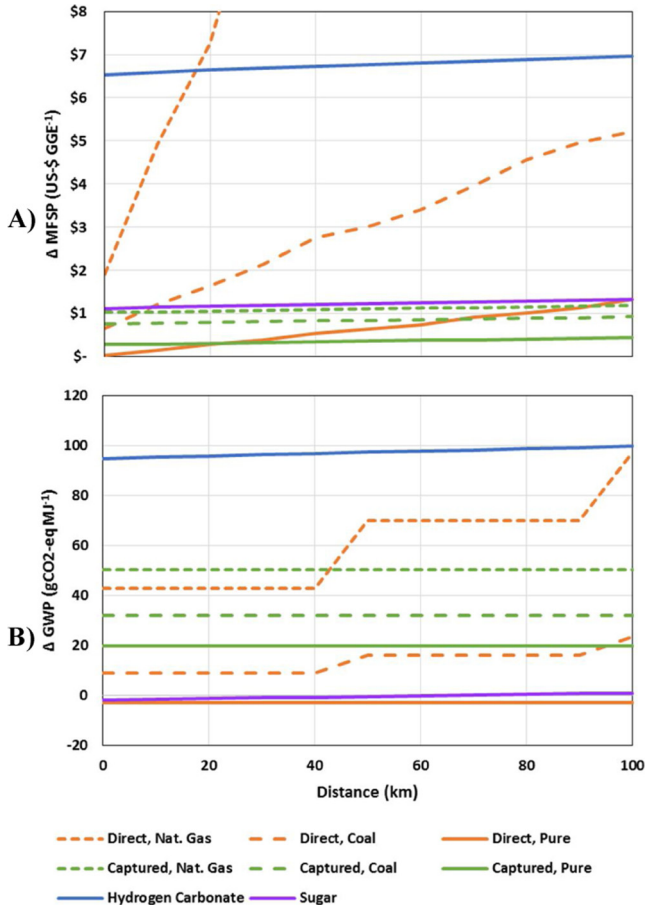

Fig. 6. Transportation distance sensitivity. (A) Minimum fuel selling price (MFSP) in 2014 US-$ per gallon of gasoline equivalent and (B) global warming potential (GWP) in grams of CO_2-equivalent per MJ of fuel for the carbon sources evaluated in this study vs. transport distance via pipeline. Reported as differences relative to the baseline MFSP and GWP of 6.47 US-$ GGE^{-1} and 28.1 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively. All cases assume 90% utilization of the carbon by the microalgae, except for hydrogen carbonate and sugar which assume 100%.

ranging from 0 to 100 km.

In general, MFSP is relatively insensitive to transportation distance, with the exception of transporting direct waste streams of CO_2 at low-pressure from power plants or pure sources. To transport the large volume of direct, dilute waste streams of CO_2 at low-pressure, multiple pipelines were required to keep the diameter and flow velocity within the design limits. Due to large diameter pipes, as well as parallel pipelines, the overall cost is a strong function of pipeline length. For all cases, costs are relatively proportional to pipeline length. It should be noted that the trends are not exactly linear due to discontinuous pipeline diameters.

A question of interest in the literature is the maximum distance over which low-pressure, direct waste streams of CO_2, like power plant flue gases, may be transported cost effectively. One way to answer this question is to determine the distance for which the direct waste stream cost matches that of transporting captured and compressed pure CO_2 100 km in a supercritical pipeline. The results from this study suggest that flue gas from a natural gas power plant cannot be transported any distance for the same cost as transporting captured and compressed CO_2 from an equivalent plant 100 km. Flue gas from a coal power plant may be transported up to 4 km, while waste CO_2 emissions from a pure source may be transported up to 40 km. These results are similar to those reported in literature. Benemann et al. [24] concluded that the maximum economical distance of CO_2 transported as low-pressure flue gas in 2-meter diameter pipes was 5 km, and Quinn et al. [22] reported 4.8 km for a similar case. In the 2016 Billion-Ton Report published by the U.S. Department of Energy, it was reported that for a natural gas power plant, coal power plant, and ethanol plant, cost-effective distances for CO_2 transport are less than 0.8 km (0.5 miles), 8 km (5 miles), and greater than 32.2 km (20 miles), respectively [16]. The consistency of literature and the results from this study reinforces the conclusion that transport of low-pressure flue gas is limited to short distances.

Similar to MFSP, GWP is relatively insensitive to transportation distance, with the exception of transporting direct waste streams of CO_2 at low-pressure from power plants. Due to significant losses, booster stations (equally spaced) are required for those pipelines greater than 40 km, which can be seen in Fig. 6 as step increases in GWP. It should be noted again that there is no difference between co-location and transport cases (under 100 km) for captured and compressed CO_2 as it must still be compressed on-site to be temporarily stored as a liquid. These results demonstrate the significant challenge and energy required to transport CO_2 in a gaseous state over great distance.

3.6. Sensitivity to carbon utilization efficiency

Unless CO_2 is directly supplied to open raceway ponds dissolved in water, gaseous losses to the atmosphere are expected. As CO_2 utilization efficiency decreases, a greater volume of CO_2 supply is needed. This increase in volume results in additional resource cost and energy, so there is incentive to maximize utilization efficiency. However, to achieve high utilization efficiency, expensive technologies and control strategies may be required, so it is important for process designers to understand the tradeoffs between sustainability metrics and CO_2 utilization. Carbon utilization efficiencies reported in literature are wide-ranging; [1] reported a utilization efficiency of 26% for an internal loop airlift reactor, while the Aquatic Species Program reported greater than 90% utilization efficiency in an outdoor open-raceway pond through counterflow injection of CO_2 [44]. The risk is that estimates for carbon utilization efficiency have been used in sustainability assessments without regard to their actual accuracy. The original sources and designs to achieve a given utilization are often not considered and therefore no longer accurately represented in theoretical assessments. This knowledge gap could have significant implications on the economic and environmental impacts of carbon delivery and presents an opportunity for future research.

For the purposes of this study, sensitivity to carbon utilization efficiency was considered. Presented in Fig. 7 are model results of MFSP and GWP for all gaseous CO_2 cases (excluding atmosphere) as a function of utilization efficiency. In general, direct waste CO_2 is less sensitive than captured and compressed CO_2, and to some extent, all cases increase nonlinearly as utilization efficiency decreases. The least sensitive cases are direct waste CO_2 from coal power plants or pure sources, for which 70–80% or 20–30% utilization efficiency may be acceptable with little penalty, respectively. This is due to low energy requirements and operating costs. All other case results indicate a need for greater than 90% utilization efficiency without incurring significant penalties to MFSP and GWP. Captured and compressed CO_2 is more sensitive than direct waste emissions due to the significant energy requirements and added operating costs. Direct waste emissions are more sensitive for natural gas power plants than coal power plants or pure sources because of the significant increase in gaseous volume. These results demonstrate the importance of understanding and not overestimating the carbon utilization efficiency.

3.7. Resource considerations

For the purposes of this study, the sustainability of carbon delivery was evaluated assuming annual average algal productivity values. In practice, however, algal productivity will be variable. Davis et al. [26] reported an annual average of 25 g m^{-2} day^{-1} but indicated that the

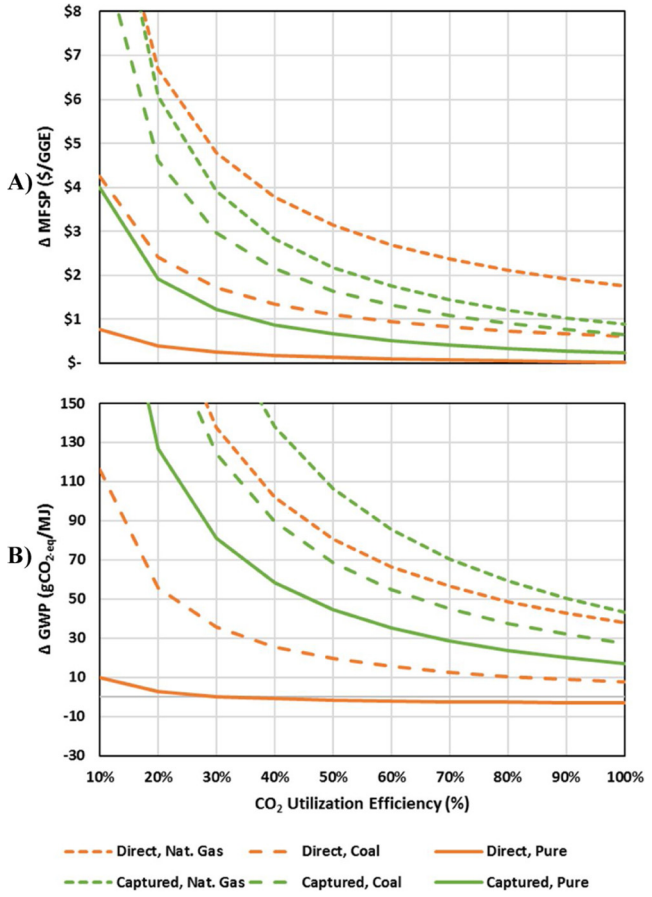

Fig. 7. Carbon Utilization Efficiency Sensitivity. (A) Minimum fuel selling price (MFSP) in 2014 US-$ per gallon of gasoline equivalent and (B) global warming potential (GWP) in grams of CO_2-equivalent per MJ of fuel for the carbon sources evaluated in this study vs. CO_2 utilization efficiency. Reported as differences relative to the baseline MFSP and GWP of 6.47 US-$ GGE^{-1} and 28.1 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively. All cases assume co-location with the microalgae facility.

summer peak and winter low would be 35 and 11.7 g m^{-2} day^{-1}, respectively. Designing a facility to manage the variability of production throughout the year can be accomplished in one of two ways: design and size the facility for summer peak productivity at the cost of underutilization during the winter or design the facility for an average productivity and include biomass and resource storage to offset extra growth during the summer peak period and maintain production yields. The latter is preferred as it minimizes equipment and costs, but material storage is a challenge. Productivity may also vary on an hourly and daily basis, which poses a significant challenge to carbon delivery. Photosynthetic algae can only grow and utilize CO_2 during the daylight hours, but CO_2 sources like power plant emissions are available 24 hours a day. This suggests the need for adequate carbon storage so that an algal facility may be decoupled from its source of carbon and provide a reliable carbon sink for industries considering algae as a carbon capture and utilization strategy. Of the sources considered in this study, only captured and compressed CO_2, hydrogen carbonate and cellulosic sugar could be sustainably stored. In general, further research is required to fully understand the implications of variable productivity on the sustainability of carbon delivery and resource assessment.

The results from this study may be used to better inform detailed resource assessments, but for the purposes of demonstration, a basic resource assessment was investigated. According to 2016 data from the EPA, 1911 million metric tons (Mt) of CO_2 is emitted every year by 1816 point sources of CO_2 emissions [62] in the United States. The point sources considered include coal and natural gas power plants, fertilizer (ammonia), hydrogen, and ethanol plants. A map of these point sources in the continental US may be found in the supplemental material. It should be noted that the EPA database includes only sources that emit greater than 25,000 metric tons of CO_2 per year. Of the 1911 Mt CO_2 $year^{-1}$ estimate, 65.6%, 29.6%, 1.5%, 2.3%, and 1% of the CO_2 emissions are from coal power plants, natural gas power plants, ammonia, hydrogen, and ethanol plants, respectively. The results from this study suggest that coal and natural gas power plants are not sustainable sources, so those may be eliminated. Furthermore, the highest potential productivity for microalgae within the continental United States may be achieved in southern California, Arizona, New Mexico, and west Texas [22]. With those constraints applied, only 20 of the 1816 sources remain, which comprise less than 0.5% of the potential CO_2 resource to be captured in microalgae and converted to fuel. Assuming 30% fuel yield, 90% CO_2 utilization, and neglecting land restrictions, this results in a maximum potential of approximately 1363 million liters (360 million gallons) of fuel per year. Though the potential for fuel production is significant, the potential to capture and recycle CO_2 emissions from existing point sources is not. While this assessment is limited, it does highlight the need for a more detailed, geographically resolved assessment leveraging the results from this study.

4. Conclusions

In this study, carbon delivery was considered for an algal biorefinery and further evaluated for sustainability. The carbon sources evaluated were CO_2 (atmospheric, direct chemical or power plant waste emissions, and concentrated), hydrogen carbonate salt, and cellulosic sugars, and evaluated both co-located and transported by pipeline. It was found that the economics of carbon delivery are more prohibitive to a sustainable system than energy and emissions. The only two cases to meet both the economic and environmental criteria of contributing less than 0.50 US-$ GGE^{-1} and 20 g $CO_{2\text{-eq}}$ MJ^{-1} to the system, respectively, were uncompressed, pure sources of gaseous CO_2 with pipeline transportation of 40 km or less; and compressed, supercritical CO_2 from pure sources for pipeline transportation up to 100 km. The scalability of algal biofuels based on these results show carbon to be a limiting nutrient in an algal biorefinery. Carbon delivery is often overlooked in sustainability assessments, but this study demonstrates its importance. The results of this research may be used by the algal research community to inform other techno-economic assessments, life-cycle assessments, resource assessments, and new process designs to better evaluate the impacts and define the requirements of carbon source selection for a sustainable system.

Declarations of interest

None.

Acknowledgments

This work was completed as part of *A Novel Platform for Algal Biomass Production Using Cellulosic Mixotrophy* project led by Arizona State University and in collaboration with the National Renewable Energy Laboratory and the U.S. Department of Energy (DOE Award Number DE-EE0007562). The authors would like to acknowledge Evan Sproul and Jesse R. Cruce of Colorado State University for editing the manuscript.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.jcou.2019.01.007.

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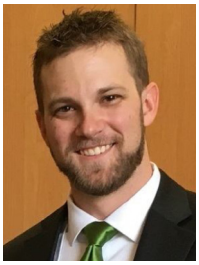

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